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Nadia Elmasry and S. M. Bedair

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Ferromagnetic Properties of GaMnN and Charge Transfer Carriers at Hetero-interfaces

Principal Investigators

N. A. El-Masry and S. M. Bedair
North Carolina State University
Raleigh NC 27695

Abstract

We report on the dependence of ferromagnetic properties of MOCVD grown GaMnN films on carrier transfer across adjacent layers. We found that the magnetic properties of GaMnN, as a part of GaMnN/GaN:Mg heterostructures depends on the thickness of both the GaMnN film and the adjacent GaN:Mg layer and on the presence of a wide bandgap barrier at this interface. These results are explained based on the occupancy of the Mn energy band and how the occupancy can be altered due to carrier transfer at the GaMnN/GaN:Mg interfaces.

Diluted Magnetic Semicondutors (DMSs) are potential building blocks for devices that make use of the spin degree of freedom of charge carriers. Recently Mn-doped III-V DMS materials have attracted a lot of attention due to the presence of ferromagnetic ordering [1]. Munekata *et al*. [2] demonstrated that mediation of ferromagnetic properties could be turned on and off by photo-generation of carriers in the InMnAs layer of an InMnAs/GaSb heterostructure. This is a clear indication that DMS properties show a dependence on the presence of carriers in the ferromagnetic layers. Room temperature ferromagnetism in GaMnN material system has been demonstrated by several groups [3-5]. It has been theoretically predicted that the ferromagnetic properties observed in the GaMnN material system depends on the occupancy of the Mn energy band in GaMnN films and the position of the Fermi level (E_f) relative to this band [6-7]. Carriers (holes) in the Mn energy band are needed to mediate ferromagnetism; the depletion and enhancement of carriers in this band will change the ferromagnetic properties of GaMnN films. This concept will be demonstrated in this work, by studying the carrier transfer across the GaMnN/GaN:Mg interface, which results in a change in the occupancy of the Mn energy band, hence affect the ferromagnetic properties of the GaMnN films.

In this study GaMnN films were grown by Metalorganic Chemical Vapor Deposition (MOCVD) on (0001) sapphire substrates using TMGa, TMAl, $(EtCp)_2Mn$ and Cp_2Mg as organometallic precursors. Ammonia and silane were used as active nitrogen and silicon sources, respectively. A typical growth run starts by depositing a low temperature (LT) GaN layer followed by ramping the temperature to 1000°C to deposit a ~1.5μm undoped layer of GaN. This is the basic structure common to all films in this study. Details of the layers deposited on this template are discussed below.

Magnetic characterization of the films was performed at room temperature by a Superconducting Quantum Interference Device (SQUID) magnetometer. Secondary Ion Mass Spectroscopy (SIMS) was utilized to determine the concentration of both Mn and Mg atoms in the grown films.

Ferromagnetic GaMnN is grown by MOCVD and DMS properties are observed at room temperature. The Mn concentration in these films is in the range of 0.1 to 0.3% which is and order of magnitude lower than that reported by MBE grown GaMnN. However, in the current study the starting GaMnN films have been grown at a temperature, and other growth parameters, that are outside of the range of parameters that lead to ferromagnetic GaMnN films. The Mn concentration in all of these films was found to be 10^{20}atoms/cm^3 as measured by SIMS.

It is demonstrated in this study that the very weak ferromagnetic or paramagnetic GaMnN films can be rendered ferromagnetic by changing the occupancy of the Mn energy band in the GaMnN layer by utilizing it as a part of several structures. Structures that are used in this study are labeled A, B and C. Mg concentration, as measured by SIMS, is 5×10^{19} atoms/cm^3 in these structures.

Structure A is a GaMnN/GaN:Mg single heterostructure (SHS) in which the GaN:Mg layer thickness is varied from 0.15 to 0.75 µm, whereas the GaMnN layer thickness was kept constant at 0.375 µm. Figure 1 (a) shows the initial weak ferromagnetic behavior of the GaMnN/GaN:Mg heterostructure, and how the presence of the GaN:Mg film changes the magnetic properties. The magnetization response from these structures scaled with the GaN:Mg layer thickness and eventually saturated as shown in Figure 1 (b).

Structure B comprises of a GaMnN/GaN:Mg multi layer double heterostructures (DHS) that are repeated three times. The GaN:Mg layers were kept at a fixed thickness of 0.125 µm, whereas the GaMnN thickness was varied from 0.125 to 0.2 µm. It is shown in Figure 2 (a) that ferromagnetism is only observed in GaMnN films with $t_a > 0.162$ µm. Figure 2(b) shows the paramagnetic and ferromagnetic responses of these structures with 0.125 and 0.2 µm GaMnN layers.

Structure C are GaN:Mg/AlGaN/GaMnN/AlGaN/GaN:Mg multilayer structures with the AlGaN layers serving as a wide bandgap barrier for the carriers. The thickness and composition of the GaN:Mg (0.75 µm) and GaMnN (0.38 µm) layers were chosen such that the resulting films would be ferromagnetic in the absence of the AlGaN barriers, based on the data presented in Figures 1 and 2. The AlGaN film thickness, t_b, varied from 25 to 200 nm and the Al concentration was kept at 30%. The magnetic properties of the multilayer structure are shown in Figure 3 (a). The SQUID data indicate that the GaMnN films are ferromagnetic only for very thin AlGaN barriers ($t_b \leq 25$nm) and are slightly ferromagnetic for thicker AlGaN barriers as shown in Figure 3(b).

In order to explain our results we should first discuss the difference of the behavior of Mn atoms in GaMnAs and GaMnN compounds. For GaMnAs, Mn is a relatively shallow acceptor which generally substitutes for Ga atoms in the lattice providing localized magnetic moments and holes [1]. Thus the abundance of holes in the valence band that exists in this p-type GaMnAs mediates the magnetic interaction.

For GaMnN the situation is quite different. While the Mn atoms still substitute for Ga atoms in the lattice, they act as deep levels in GaN and form an impurity energy band whose width depends on the Mn concentration [7-8]. Optical absorption measurements indicated the Mn energy band (E_{Mn}) is located 1.4 eV above the valence band edge (E_{V}) of GaN [8]. The interaction of the Mn energy band with the valence band is very small and it is not expected to polarize the valence band. Thus, carrier mediated ferromagnetism and 100% spin-polarized band in GaMnN can only be present if the E_f resides within the Mn energy band. Therefore, the location of the E_f will determine the occupancy of the Density of States (DOS) in this impurity band and thus the availability of carriers to mediate ferromagnetism.

The current results can be explained based on the carrier transfer from the Mn energy band to the Mg level and its effect on the occupancy of the Mn energy band. The Mn energy band in GaMnN and the Mg level in GaN:Mg are located about 1.4 and 0.15 eV above the valance band of GaN respectively as shown by the schematic in Figures 4 (a) and 4(b).

At room temperature, the Mg acceptor level in GaN:Mg is barely activated, only ~1% activated. Therefore, for a Mg concentration of $5x10^{19}/cm^3$, the hole concentration in the valence band is in the low $10^{17}/cm^3$ range. Thus the GaN:Mg has an acceptor level with concentration in $10^{19}/cm^3$ range. We will initially assume that the Mn energy band is completely filled with electrons; this assumption will be addressed later. Thus for the GaMnN/GaN:Mg SHS (structure A) electrons in the completely filled Mn energy band of the GaMnN layer will transfer into the acceptor Mg level in the adjacent layer, resulting in a partially filled Mn energy band. This partially filled Mn energy band will offer the required conditions to mediate ferromagnetism in these GaMnN/GaN:Mg SHS. Therefore these films will be ferromagnetic with the magnitude of the magnetization depending on the availability of Mg acceptor levels and in turn the thickness of the GaN:Mg layer as shown in Fig 1(b). The carrier transfer process will continue until a built in electric field and a depletion layer are established at the GaMnN/GaN:Mg interface. There will be two consequences of this diffusion process. The first is the partial emptying of the Mn energy band to yield the desired carriers and hence the mediation of ferromagnetic properties. The second is the formation of a depletion region at the interface and a built in electric field.

For the GaMnN/GaN:Mg multi layer DHS (structure B) in which ferromagnetism was only observed for GaMnN films of t_a>0.162 μm (fig2(a)). Very thin GaMnN films are depleted from carriers and these films show paramagnetic responses. However, these layers do not suffer from carrier depletion throughout their whole thickness when they are grown thicker and are able to offer ferromagnetic properties as shown by curve (x) of Figure 2 (b).

For the GaN:Mg/AlGaN/GaMnN/AlGaN/GaN:Mg DHS (structure C), the saturation magnetization was very weak except for AlGaN barrier layer less than 25 nm in thickness. The presence of the AlGaN barrier layers will affect the carrier transfer from GaMnN to the GaN:Mg layers. Carrier transfer across the barrier will take place either by tunneling or thermionic emission. The probability of tunneling will exponentially decay with the AlGaN barrier thickness (t_b). For very thin AlGaN barriers (t_b <25nm) the charge transfer across the GaMnN/GaN:Mg interface is not impeded and ferromagnetic properties are retained. However for t_b>50nm, the charge transfer across the GaMnN/GaN:Mg interface is highly impeded and the ferromagnetic response is extremely weak or the films are practically paramagnetic. The dependence of magnetization on t_b and the hysterisis curves for AlGaN barrier thicknesses of 25 and 50 nm is illustrated in Figure 3(a).

The discussion above was based on the assumption that the E_f is located above the Mn energy band and that this band is filled with carriers. To check this assumption, the starting non-ferromagnetic GaMnN layer was grown between GaN:Si layers to form a GaN:Si/GaMnN/GaN:Si DHS and it was found to be very weakly ferromagnetic. The data can be explained as follows: Si is a shallow donor in GaN with an energy level several meV below Ec. Therefore if the E_f in GaMnN film is close to Ev, the Mn energy band will be empty. Electrons from the GaN:Si layers will transfer (diffuse) to the GaMnN layer and result in a partially filled Mn energy band. Thus ferromagnetic properties should be observed. Since this is not the case in the experimental findings, the initial assumption that GaMnN has a completely filled Mn energy band is valid. Also if the Mn energy band was empty, the presence of a neighboring Mg level would not generate the required carriers to mediate ferromagnetism.

In conclusion the above results reveal several important aspects of ferromagnetism in GaMnN. It is suggested that ferromagnetism in GaMnN is dependent on the availability of carriers in the Mn energy band to mediate ferromagnetism. The availability of these carriers depend on the position of the E_f in GaMnN films. In this study our starting film is not ferromagnetic, since the Mn energy band is completely filled with electrons. Ferromagnetic properties can be induced in this paramagnetic GaMnN film by partially depleting electrons from the Mn energy band. The data presented should not be evaluated from a quantitative point of view, but rather should be treated as a qualitative trend. There are several unknown parameters that will make quantitative analysis difficult to present such as the exact location of the Mn energy band, the width of this band and the presence of other impurities that can affect the position of E_f. However, even with such qualitative discussions, a consistent set of results can allow better understanding of the ferromagnetic properties of GaMnN. Thus it is fair to conclude that ferromagnetism in MOCVD grown GaMnN is carrier mediated within the Mn composition range used in this study.

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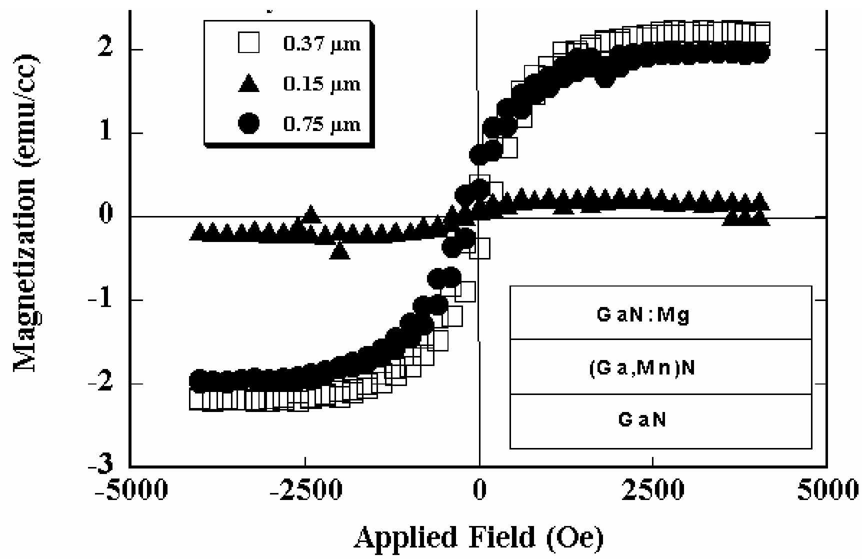


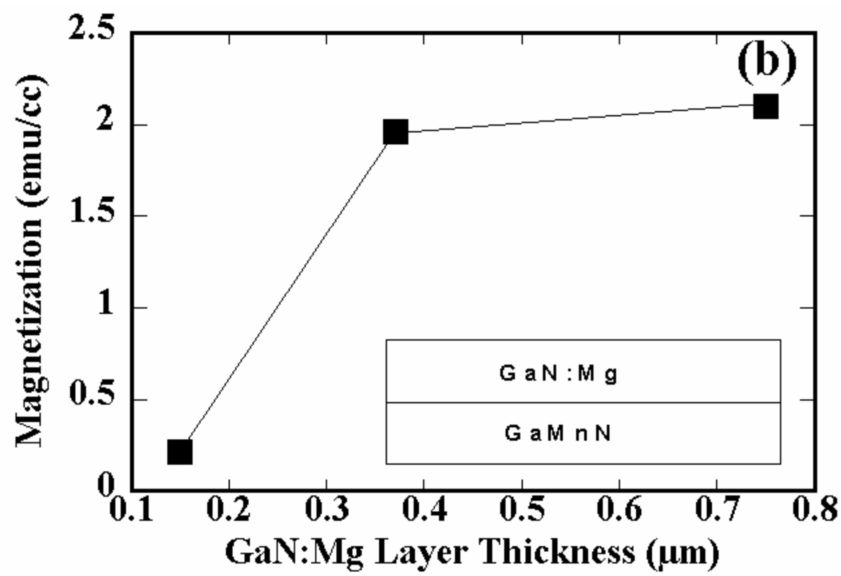
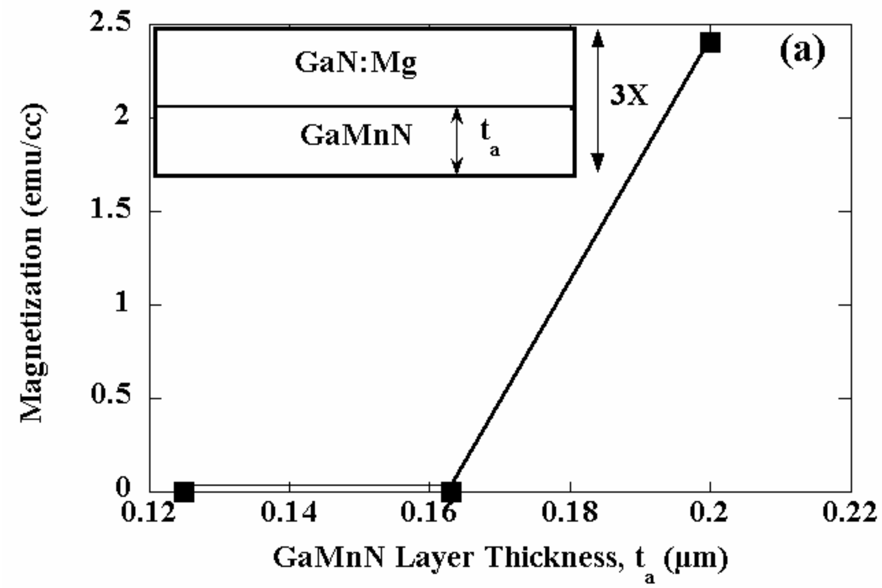


Figure 1: (a) Magnetization vs. applied field curve for structure A. ▲ very weak ferromagnetic response of GaMnN with 0.15µm GaN:Mg grown on top. ߐ and ● are ferromagnetic responses for GaN:Mg film thickness of 0.37 µm and 0.75 µm

respectively. (b) Change in magnetization as a function of GaN:Mg layer thickness for structure A.



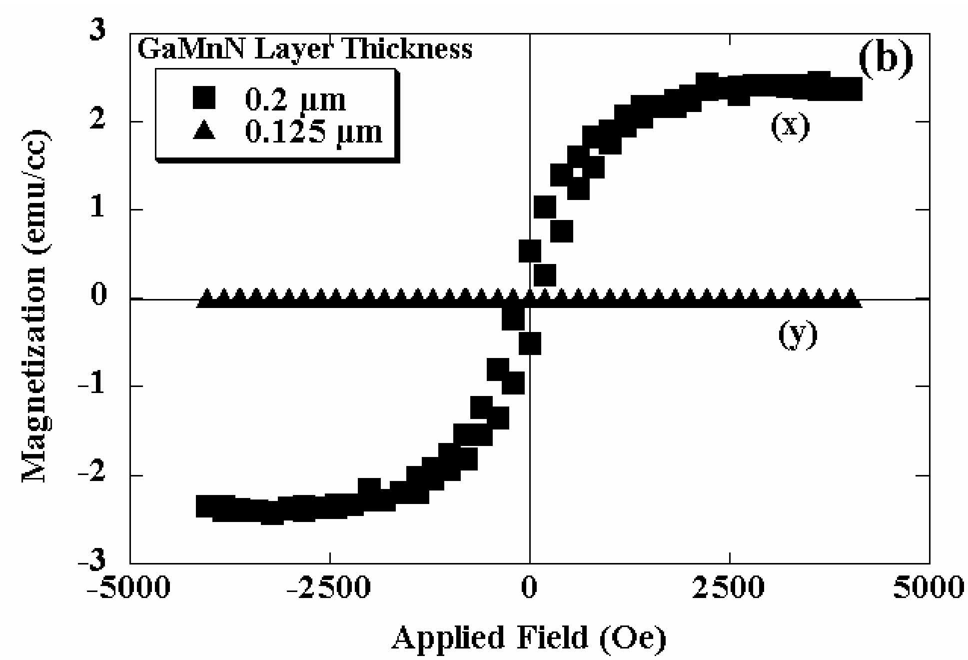


Figure 2: (a) Magnetization vs. GaMnN layer thickness t_a. GaMnN/GaN:Mg multi layer DHS as shown in inset. At room temperature ferromagnetic properties are only observed for GaMnN films thicker than 0.162µm. (b) Magnetization vs. applied field showing the

change in ferromagnetic properties as the thickness of the GaMnN layer is increased from 0.125μm (curve y) to 0.2μm (curve x).

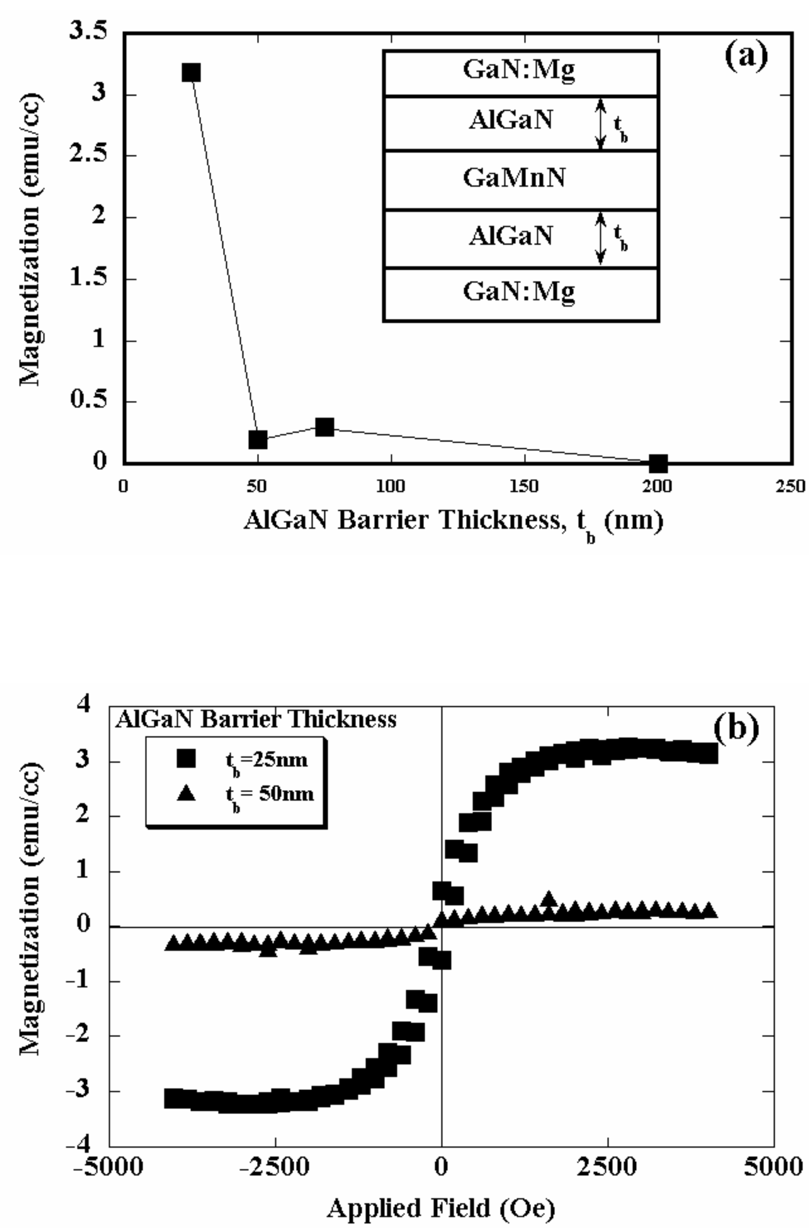


Figure 3: (a) Magnetization vs. AlGaN barrier thickness for GaN:Mg/AlGaN/GaMnN/AlGaN/GaN:Mg DHS The thickness of the GaMnN and GaN:Mg layer are fixed at 0.38 and 0.75μm, respectively. (b) Magnetization vs. applied field for DHS containing 25 and 50nm thick AlGaN barriers.

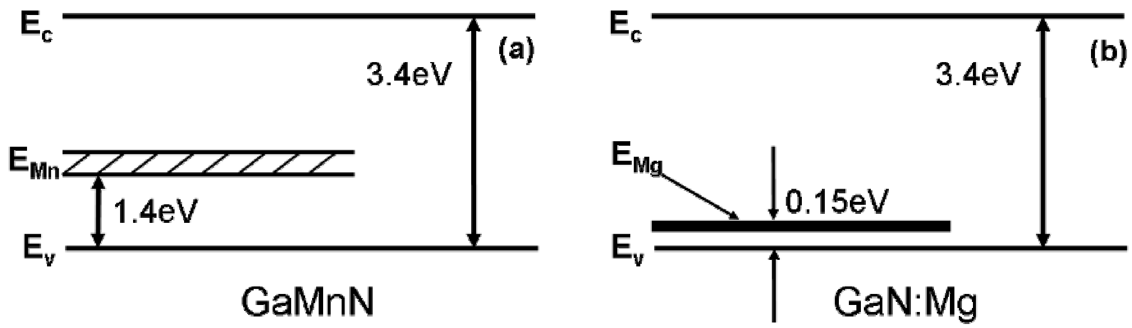


Figure 4: Schematic of band diagrams showing different energy bands and levels for (a) GaMnN (b) GaN:Mg.